Preparation and Certification of Performance Evaluation Materials: Pu-239 in Artificial Urine at the μBq (~100 aCi) Level

A Work in Progress

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I. Introduction

The Department of Energy is in the process of assisting Marshall Island residents to resettle their formerly inhabited islands after five decades. The DOE must ensure that the residents, when returned to their native islands, are not unnecessarily exposed to radiation from the intake of residual radioactivity in the environment. The DOE has various ongoing environmental monitoring and radiobioassay programs that will be utilized to ensure that the radiation dose to residents will not exceed recognized international standards. One of the remaining radionuclides giving rise to an internal radiation dose from inhalation and ingestion intake pathways is ^{239}Pu. The uptake of ^{239}Pu in the body from all pathways can be estimated through the quantification of the daily excretion of ^{239}Pu in the urine of an individual. For this application, the radiobioassay techniques must be extremely sensitive and have a detection level several orders of magnitude below the more routine alpha spectrometric method employed by most laboratories for environmental monitoring and occupational radiobioassay program applications.

I. a. Current Radioanalytical Methodology

During the last decade, a technique having the necessary sensitivity to evaluate ^{239}Pu at the μBq/L level was developed at the Brookhaven National Laboratory (1,2). This technique involves the radiochemical purification and concentration of Pu in the urine specimen followed by the evaporation of the final μL analyte solution on a fission track detector and exposure to neutrons in a reactor. This technique has been refined over the years and has been routinely employed as the radiobioassay technique for the DOE's Marshall Island program. Although this technique has been found to be very suitable and essentially the only method available, it has certain drawbacks. The method would be characterized as being relatively complex and requires extreme contamination control precautions, from both a radiochemically and clean room/facility point-of-view. In addition, the technique is rather expensive when viewed on a cost per sample basis and is inherently time consuming due to its complexity and the requirement of a neutron source.

I. b. Newly Developed Technique Under Evaluation

During the past five years, a highly sensitive and relatively less expensive method involving inductively coupled plasma - mass spectrometry (ICP-MS) has been developed for environmental monitoring, site remediation and radiobioassay applications involving long-lived radionuclides (3,4). Several well-known laboratories have adapted the basic ICP-MS method to the measurement of ^{239}Pu in bioassay specimens at concentration levels that are comparable to those attained by the fission track analysis method. The ICP-MS technique, when compared to the fission track analysis method, is potentially more quantitative, less expensive, simpler, quicker, not susceptible to contaminants and does not require the availability of a neutron source such as a reactor. Both techniques require the up-front radiochemical processing of the urine specimen for purification and concentrating the analyte.

II. Broad Objectives

In the near future DOE intends to implement a radiobioassay program for the resettling residents of the Marshall Islands that employs the most reliable, cost effective and sensitive technique available. To this end, the existing high sensitivity methodologies must be evaluated, validated and compared under the same test conditions. Quality and performance parameters (such as sensitivity, precision, accuracy and representativeness) of the techniques at μBq levels will be evaluated in this study.

The broad objectives of this work can be summarized as the following:

- demonstrate the protocol for the preparation of SRMs having μBq levels ^{239}Pu in artificial urine that will be used as proficiency examination samples in national quality control programs,
- validate the fission track analysis method for μBq levels of ^{239}Pu in urine samples,
- validate the ICP-MS method for μBq levels of ^{239}Pu in urine samples,
- document the performance capability and comparison of the fission track analysis and ICP-MS techniques,
- select laboratories to conduct routine, low-level ^{239}Pu bioassay programs for Marshall Island resettlement and
- provide the technical justification for potential savings of federal government funds.

III. Specific Technical Objectives

The National Institute of Standards and Technology and the Yankee Atomic Environmental Laboratory have collaborated to jointly develop and implement a SRM/PE sample preparation protocol. Once certified, these μBq level samples will be used to conduct a round robin testing program that will evaluate and compare the fission track and ICP-MS methodologies.

The specific technical objectives of this work can be summarized as:

- develop a SRM/PE sample preparation protocol that is cost effective, technically sound and defensible,
- prepare SRM/PE materials for four ^{239}Pu concentration levels, four laboratories, five samples per concentration level including blanks,
- document all gravimetrically and radiometrically determined dilutions, final quantification and total uncertainty of each SRM/PE sample prepared,
- provide project documentation and quality assurance assessments conducted by NIST for the purpose of preparing the certificate of content for the SRM/PE samples,
- conduct round robin testing of participating laboratories, and
- statistically evaluate the test results for bias, precision and methodology.

Since the project will commence during the fourth calendar quarter of 1996, this presentation will be considered as a "work in progress." The protocol to be used in the preparation of the SRM's and various aspects of the project that will have been completed by the time of the conference will be presented.